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GENERAL MOTORS CORPORATION

TECHNICAL REPORT

ON

THE DIAGNOSTIC APPLICATIONS OF MICROWAVE ABSORPTION AND DISPERSION

Sponsored By

ADVANCED RESEARCH PROJECTS AGENCY

Monitored By

U.S. ARMY MISSILE COMMAND

CONTRACT NO. DA-01-021-AMC-11359(Z)

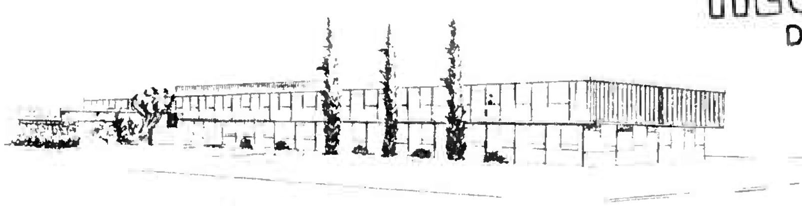
HYPERVELOCITY RANGE RESEARCH PROGRAM

A PART OF PROJECT "DEFENDER"

GM DEFENSE RESEARCH LABORATORIES

SANTA BARBARA, CALIFORNIA

AEROSPACE OPERATIONS DEPARTMENT



TR65-01D

JANUARY 1965

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TECHNICAL REPORT

ON

THE DIAGNOSTIC APPLICATIONS OF MICROWAVE ABSORPTION AND DISPERSION

I. P. FRENCH

THIS RESEARCH WAS SUPPORTED BY THE
ADVANCED RESEARCH PROJECTS AGENCY
AND WAS MONITORED BY THE
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REDSTONE ARSENAL, ALABAMA

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FOREWORD

This report is one of a series of related papers covering various aspects of a broad program to investigate the flow-field variables associated with hypersonic-velocity projectiles in free flight under controlled environmental conditions. The experimental research is being conducted in the Flight Physics Range of GM Defense Research Laboratories, General Motors Corporation, and is supported by the Advanced Research Projects Agency under Contract No. DA-01-021-AMC-11359(Z). It is intended that this series of reports, when completed, will provide a background of knowledge of the phenomena involved in the basic study and thus aid in a better understanding of the data obtained in the investigation.

The present report is one of a group of three related reports within the above series, all with the same author and treating various aspects of the same subject. The two other reports in this group are:

TR65-01A, "A Review of Microwave Absorption and Dispersion of Air Constituents," and

TR65-01C, "The Microwave Absorption and Dispersion of Heated Air."

The work described in this group of reports was supported partly under the basic contract but includes work supported by GM DRL with in-house funding.

ABSTRACT

The possibility of measuring species concentration in high-temperature air by microwave spectroscopy is considered. The species selected for consideration are molecular oxygen, which has transitions in the region from 52 to 68 Gc, and nitric oxide with transitions around 150 Gc. The feasibility appears marginal, but this evaluation is based on many approximations and assumptions and further work is required for definitive conclusions.

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ILLUSTRATIONS

I. INTRODUCTION

The current interest in hypervelocity flight has stimulated a great deal of investigation on the dissociated and ionized plasma sheath and wake around a hypervelocity vehicle. In particular it is very desirable to be able to predict the interaction of electromagnetic waves with this ionized environment. This cannot be done unless the magnitude of electron density, which is closely related to the concentrations of the other constituents in heated air, is known. In addition, the collision frequency of electrons with other particles and the spatial variation of all the constituents is required. Measurements on full-scale hypervelocity vehicles are expensive and difficult, so laboratory facilities for simulating various aspects of hypervelocity flight have been used quite extensively in recent years. One type of facility is the hypervelocity range,[(1)*] where small models are fired at high velocity into a controlled environment and measurements on the flow field and wake are made. Another very useful tool for studying the chemical kinetics of reactions is the shock tube,[(2)] convenient because aerodynamic considerations are largely unimportant in this essentially one-dimensional flow.

Among the instruments used for measuring electron density in these facilities are electromagnetic probes, using either optical[(3)] or microwave frequencies.[(4)] These instruments measure the effective refractive index of the heated, ionized gas as it goes by. The electron density is then deduced from the refractive index. However, the atoms and molecules (which may be neutral) also have an effective index of refraction different from unity. The two effects have very different dispersive behavior, so that it is possible in principle to separate out the two effects by using different frequencies. In fact, it appears possible to measure both the electron density and the concentration of some particular species in the flow by this method.

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* References are listed at the end of this report.

It would be most desirable to measure the concentrations of some species in the far wake, because there are a number of predictions (for example, References 5 and 6) of what the magnitudes ought to be. These predictions could be compared against experiment to deduce more accurate values of the rate constants for the various chemical reactions.

Of particular interest is the feasibility of using a microwave Fabry-Perot cavity[7, 8] to measure the concentration of some species (such as molecular oxygen) in the wake of hypervelocity projectiles in a free-flight range. It appears possible to single out one of the many species in the flow by using a probing frequency in the vicinity of a natural resonance frequency of the species under investigation. It is well known[9] that the refractive index (assumed complex) of a gas is highly dispersive around a resonance, so that hopefully the effects of the other species in the flow would be negligible in comparison.

The complex dielectric at $1,000^{\circ}K$ was examined in References 10 and 11, where the contribution of molecular oxygen transitions in the frequency range from 52 to 58 Gc was discussed in detail. Also considered was the band of nitric oxide lines around 150 Gc. Nitric oxide was considered because of its importance in high-temperature chemical reactions in air. The results contained in References 10 and 11 will now be used to determine the feasibility of using the absorption and dispersion properties of heated air as a diagnostic technique.

II. THE FABRY-PEROT CAVITY

Rotational molecular transitions, which give rise to microwave and far-infrared spectra of gases, are generally very weak. Long path lengths of the gas must be used to increase sensitivities. Another way of increasing sensitivity is by increasing the number of times the microwave energy passes through the gas. This is done by using a microwave resonant cavity. The performance of a cavity is closely related to its "Q" value, which is given by[12]

$$Q = f/\Delta f \tag{1}$$

where f is the operating frequency and Δ_f is the linewidth of the resonance at the half-power points.

A high Q is required for the greatest sensitivity. It appears that the highest Q's are attainable with a Fabry-Perot cavity. These instruments have been developed within the last few years for millimeter wavelengths by Culshaw[13, 14] and have been applied to millimeter wave absorption spectrometry by a group at the National Bureau of Standards.[15, 16] Currently the Fabry-Perot cavity is being applied to plasma diagnostics[7, 8] at GM Defense Research Laboratories.

The Q of a Fabry-Perot cavity is given by the well-known expression[15]

$$Q = \frac{k\,L}{A} = \frac{2\,\pi\,L}{\lambda A} \tag{2}$$

where k is the wave number

L is the plate separation

and A are the cavity losses.

For an empty cavity with diffraction losses small compared to reflection losses, $A = 1 - |\Gamma|^2$, where Γ is the reflection coefficient of the reflectors. Q's of about 300,000 have been achieved recently with 10-inch 70-Gc cavities operating in a mode which is close to confocal.[8]

By using a high Q cavity, very long effective path lengths (the equivalent absorption path length in free space) can be achieved.

L_{eff} is given by[12]

$$L_{eff} = \frac{Q\lambda}{2\pi} \tag{3}$$

Consequently

$$\frac{L_{eff}}{L} = \frac{1}{A}$$

Aluminum reflection losses can be as low as 10^{-3}, giving an "improvement factor" (L_{eff}/L) of 10^3. With such a factor, Gallagher's 7-ft absorption cell[17] is equivalent to a cavity 2.13 cm long. Alternately, a two-meter cavity would have increased the sensitivity by a factor of almost one hundred. The 500-ft-long cell used at the University of Texas[18] is equivalent to a cavity 15.2 cm long. Lichtenstein[19, 20] et al have recognized that a Fabry-Perot cavity can produce this increase in sensitivity, and have measured H_2S lines at 168 Gc and at 214 Gc with a 4-inch-long cavity whose Q was 56,000. They allowed some diffraction losses to discriminate against higher-order modes, and found that lines with absorptions as low as 3×10^{-5} cm^{-1} were measurable. A longer cavity ($L \simeq 200$ cm) with a Q of greater than about 10^6 would allow one to measure lines with absorptions lower than 10^{-6} cm^{-1}.

III. USE OF A FABRY-PEROT CAVITY TO MEASURE SPECIES CONCENTRATION IN HEATED AIR

In many reentry physics experiments it would be very helpful to be able to measure the concentration of various species in shock-heated gases. Our particular interest is the Flight Physics Range at GM Defense Research Laboratories.[4] In this facility 1/2" or 7/8" models are fired at high velocity (~20,000 feet per sec) into air at low pressure (~10 mm Hg) to simulate certain aspects of hypervelocity flight. A plasma sheath and ionized wake are formed around the model and various microwave and radiation measurements are made to deduce the ionization and chemical history of the wake.

It appears that by using microwave absorption spectrometry with a high-Q Fabry-Perot cavity it may be possible to follow the concentration of some air species. The oxygen molecule with absorption resonances at about 60 Gc and with a single line at 118.75 Gc seems a promising candidate, and is of particular importance since generally molecular oxygen starts to reappear when the electrons disappear. Another interesting candidate is nitric oxide, whose concentration generally follows that of the electron density. As noted in previous reports[10, 11] both of these species have resonances in the millimeter-wavelength part of the spectrum and might therefore be studied with Fabry-Perot cavities.

The effect of the gas in the cavity is twofold. In the first place the gas absorption will increase cavity losses and reduce the Q. In the second place the change in the refractive index as one goes through a resonance causes a shift in the cavity resonance frequency. Both of these effects might be measured by exciting the cavity with a swept-frequency klystron. For the very high Q's under consideration ($\sim 10^6$), the cavity linewidths (~60 kc) will always be much narrower than the linewidth of the resonance it is desired to measure. The cavity resonance can then be used as a very fine probe to sweep across the line.

Let us consider the type of cavity which would be necessary to measure the 60.433 Gc oxygen line with a peak attenuation of 2 db/km or $\alpha_{\omega} = 0.23 \times 10^{-5}$ cm^{-1}. Let the cavity losses be taken as 10^{-3}, corresponding to aluminum at this frequency. The increase in Q with cavity length is linear, as shown in Figure 1 (curve a). Diffraction losses are ignored. The change in Q of a cavity resonant at 60.433 Gc when room-temperature air is introduced into it is shown by curve b. The decrease in Q is 13% for a 40-cm cavity and 25% for a 140-cm cavity. When a 4-cm-diameter wake with no oxygen molecules is introduced, however, the resulting change is very small and probably not measurable. The effect of the real part of the dielectric coefficient of air is to change the cavity resonant frequency. This frequency change, Δ_f, is given by[7]

$$\frac{\Delta f}{f} = \frac{1}{2} \frac{d}{L} \Delta K_r \tag{4}$$

where f is the operating frequency, d the wake thickness, and L the cavity length. ΔK_r is the contribution of the oxygen molecules to the real part of the dielectric coefficient. Figure 5a of Reference 10 shows ΔK_r to be $(.09 \times 10^{-4} + .706 \times 10^{-4})$ for air at 100 mm Hg pressure and 290^{O}K when the frequency is 60.156 Gc. The change in cavity resonant frequency is 2.39 Mc when the cavity is filled with air. When a 4-cm oxygen-free wake is in the cavity, the change in resonant frequency is 43 kc less. This figure is rather small when one considers that the linewidth of a $Q = 10^5$ (L = 25 cm) cavity is 600 kc. The cavity linewidth may be decreased by making the cavity longer (higher Q), but Δf due to the wake also decreases in proportion. Unless some way is found to exploit the very rapid change in ΔK with frequency, it does not appear possible to use the 60 Gc or the 118.75 Gc oxygen lines for diagnostic purposes.

We now turn our attention to the possibility of detecting the presence of nitric oxide by observing the effect of the band of lines around 150 Gc on a Fabry-Perot cavity. Table 1 of Reference 11 shows the absorption and linewidth of pure nitric oxide at temperatures of 80^{O}, 290^{O}, and 1,000^{O}K. The contribution of 1% nitric oxide to the complex dielectric coefficient is shown in Figures 6, 7, and 8

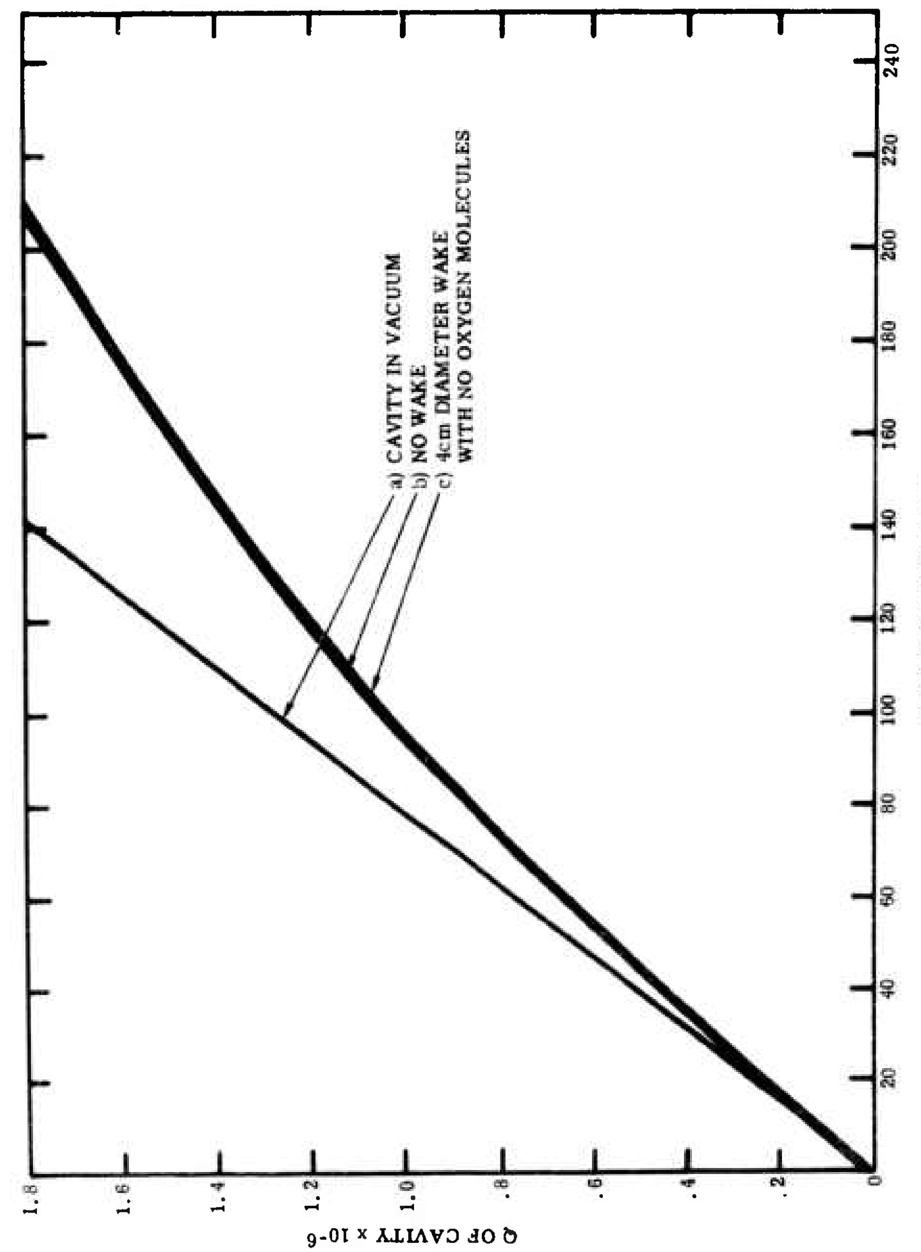

Figure 1 Effect of 60.433 Gc Oxygen Line on the Q of a Fabry-Perot Cavity. (Cavity Reflection Loss = 10^{-3} and 21% Oxygen Molecules in 290°K Air)

of Reference 11. Figure 2 of the present report shows the effect of a 4-cm 1,000°K ($p \lesssim 10$ mm Hg) wake on the Q of the Fabry-Perot cavity. The effect is very small. Ten percent of NO in the wake is shown to accentuate the change. The change in resonant Q of the cavity (Equation 4) due to $\Delta K_r = .05 \times 10^{-6}$ (Figure 7a of Reference 11) is also very small, being 600 cps when $d = 4$, $L = 25$, and $f = 150.175$ Gc. The reason Δf is so small is because the lines are much weaker at $T = 1,000^{\circ}$K than at low temperatures.

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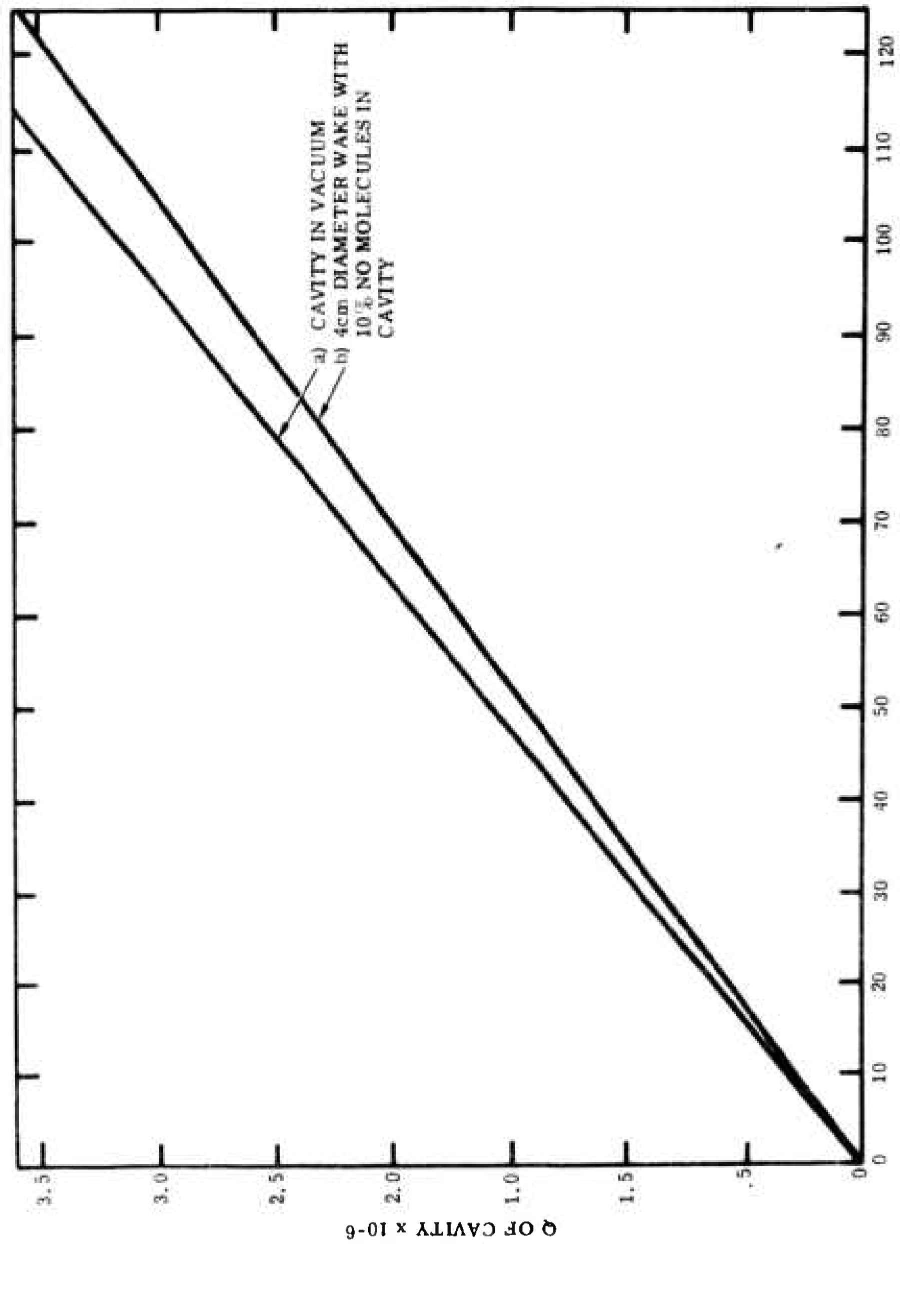

Figure 2 Effect of 150.1763 Gc Nitric Oxide Line on the Q of a Fabry-Perot Cavity (Cavity Reflection Loss = 10^{-3} and 10% Nitric Oxide Molecules in 1000°K in 4 cm Diameter Wake)

IV. THE ABSORPTION AND PHASE SHIFT CAUSED BY FREE ELECTRONS

Free electrons as well as neutral gas also have an equivalent complex dielectric coefficient different from unity, giving rise to absorption and phase shift. The power absorption α_ω and phase coefficient of free electrons when the plasma is underdense is given by[21]

$$\alpha_\omega = \frac{\nu}{2c}\left(\frac{\omega_p}{\omega}\right)^2 = \frac{\nu}{2c}\left(\frac{n}{n_c}\right) \tag{5}$$

For a frequency f of 150 Gc, the critical electron density $n_c = 2.94 \times 10^{14} \text{cm}^{-3}$. The electron collision frequency for heated air is given by[22]

$$\nu = \nu_{ea} + \nu_{ei} = 1.8 \times 10^{-8}(T/300)^{1/2} n_a + 6.1 \times 10^{-3}(T/300)^{-3/2} n_i \tag{6}$$

where n_a and n_i are the densities (in cm^{-3}) of neutral particles and ions, respectively, and T is the temperature in $^\circ$K. If we use the fact that at 290°K there are 3.33×10^{16}p molecules per cm^3 (p in mm Hg) and assume that in the heated wake of a high-velocity projectile the particle density is close to ambient, we then obtain, for an electron and ion density of $10^{10}/\text{cm}^3$,

$$\nu = 6.0 \times 10^8 (T/300)^{1/2} p + 6.1 \times 10^7 (T/300)^{-3/2} \tag{7}$$

When the temperature is 1,000°K this equation becomes

$$\nu = 1.09 \times 10^9 p$$

and the attenuation becomes

$$\alpha_\omega = 0.617 \times 10^{-16}\, n \times p$$

For $n = 10^{10} cm^{-3}$ and $p = 1$ mm Hg, $\alpha_\omega = 0.617 \times 10^{-6} cm^{-1}$. For useful microwave spectroscopy, α_ω due to the free electrons must be lower than that for oxygen and nitric oxide. Consequently, the best chance of making useful measurements is when the electron density is low as in the far wake (unfortunately, the concentration of nitric oxide will also be low under these conditions) and at the low pressures. The above value of $\alpha_\omega = 0.617 \times 10^{-6}$ cm^{-1} for free electrons compares with $\alpha_\omega = 2.70 \times 10^{-6}$ cm^{-1} (Table 1 of Reference 11) for a concentration of 1% ($n = 3.33 \times 10^{13} cm^{-3}$) 1000^oK nitric oxide in the flow. We therefore conclude that unless there are many more nitric oxide molecules than electrons in the flow it will not be possible to "see" the nitric oxide.

V. CONCLUSIONS AND RECOMMENDATIONS

We have examined the possibility of measuring concentrations of molecular oxygen and nitric oxide in the wakes of hypervelocity projectiles by detecting either the absorptive or dispersive behavior of these gases in the microwave region. The instrument under consideration for this purpose is the Fabry-Perot cavity, which can be made very sensitive. Notwithstanding this sensitivity, the possibility is marginal of making useful measurements on the lines considered. There are two reasons for this. The first is that the effect of free electrons may be larger and will swamp the effect of the molecules. The second is that although microwave transitions are easily measurable at low temperature, the intensity of the effects decreases rapidly with increasing temperature. For example, the nitric-oxide transitions decrease by at least a factor of 25 when the temperature is raised from $290^{\circ}K$ to $1,000^{\circ}K$.[11] In addition, the amount of nitric oxide in wakes is not large and is probably always less than 1%.

Recombination of atomic oxygen into molecular oxygen takes place in the far wake of hypervelocity bodies. It would be very desirable to measure the point at which this happens. The possibility is now examined of doing this by measuring the absorption of molecular oxygen as it forms. It might be just feasible to do this, but the molecular oxygen, which inevitably fills the whole cavity, may mask the effect in the wake. In a shock tube, however, the cavity could be evacuated.

The results of this study suggest that the possibility of using microwave absorption as a plasma diagnostic technique is marginal at present. However, this conclusion is based on very imperfect knowledge and very simple theoretical models. Considerably more information on subjects such as collisional broadening and how it is affected by temperature is required before definite conclusions can be drawn on the feasibility of the method in shock-tube and free-flight range studies.

Laboratory experiments are suggested as a means to greater understanding of fundamental processes, especially collisional broadening of lines and their temperature dependence.

The high Q's and narrow cavity resonances achievable by submillimeter Fabry-Perot cavities should have considerable application in the determination of spectral line shapes. This subject is of great current interest because of the fundamental information which line shapes can give on such things as interatomic and intermolecular forces and the perturbations they exert on energy levels. As noted previously (Reference 10), the positions and spacings of submillimeter lines give a great deal of information on the forces holding molecules together.

The Fabry-Perot cavity also promises to allow one to measure the anomalous behavior of the real part of the dielectric coefficient around absorption lines. As far as is known, nobody has yet measured both the real (dispersive) and imaginary (absorptive) part of the dielectric coefficient. Such measurements would allow one to check the theories of polarizability.

In conclusion, it is felt that it is very desirable to develop and extend the techniques of submillimeter-wave spectroscopy. Considerable information of basic scientific interest should be generated, and extension of these techniques may lead to useful shock-tube and free-flight range diagnostics.

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(Unclassified)

1. Microwaves – Absorption
2. Microwaves – Dispersion
3. Microwaves – Propagation
4. Microwaves – Polarization
5. Microwaves – Temperature factors
6. Dielectrics – Properties

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